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► To cite this version:

Jean-Christophe Nappé, Philippe Grosseau, Mourad Benabdesselam, Michel Beauvy, Bernard Guilhot. Characterization of aluminum nitride material under swift heavy ion irradiations. 11th International Conference of the European Ceramic Society, Jun 2009, Cracovie, Poland. pp.1105-1108. hal-00519142

HAL Id: hal-00519142

<https://hal.science/hal-00519142>

Submitted on 18 Sep 2010

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Characterization of Aluminum Nitride Material under Swift Heavy Ion Irradiations

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Abstract

For Generation IV reactors, and more particularly the Gas Fast Reactor, aluminum nitride is an interesting candidate for the application as a fuel coating; actually, its great thermal conductivity should allow efficient thermal transfer between the fuel pellets and the coolant. Unfortunately, few data are available on its behaviour under irradiation. In this study, we attempted to understand the effect of irradiation parameters on the defect creation thanks to thermally-stimulated luminescence and optical absorption spectrophotometry. Thus, we did not note any new defect created by the irradiation and the fact that these techniques complement each other allowed us to know the influence of each irradiation parameter on the defects concentration.

Keywords: AlN, irradiation, heavy ions, electronic interactions, thermoluminescence, optical absorption spectrophotometry

Introduction

In the Generation IV Forum framework, France study the Gas Fast Reactor among others, this reactor should work at high temperature (1100-1300K) and under helium gas pressure, helium being the coolant.

In order to coat the fuel, several ceramics are studied. They must meet certain specifications, such as good thermo-mechanical properties or low swelling under irradiation. Taking into account these specifications, carbides and nitrides are sensed as fuel coatings, and among these materials, aluminum nitride has one of the highest thermal conductivity: about $200 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ for a polycrystal [1-4], up to $320 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ for a single crystal [5].

However, aluminum nitride has the drawback to produce radioactive long-lived isotope ^{14}C by transmutation of ^{14}N [6]. This drawback may be reduced by the use of isotope tailored material in which ^{15}N replaces ^{14}N [7].

If the behavior of aluminum nitride under irradiation has already been studied by several authors [8-13], as far as we know no study on the defect created by irradiation has been carried out. Therefore, in this study we attempt to identify the defect induced by irradiation in aluminum nitride by thermally-stimulated luminescence and optical absorption spectrophotometry.

Experimental

Sample preparation

The studied material is a polycrystalline aluminum nitride sintered at 2123K for 3h with 3 mol% of Y_2O_3 (sintering aid). The thermal conductivity of this material was measured between 198 and $214 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. Preparation of the specimens was already described elsewhere [14]. Briefly, they were polished with diamond suspensions down to $1 \mu\text{m}$, and then subsequently irradiated with swift heavy ions.

Simulation with swift heavy ions

Heavy ions that have a kinetic energy larger than 0.1 MeV amu^{-1} are generally qualified as “swift”. This kind of ion is used essentially to simulate the impact of fission products. However, by studying the spoiling at the end of the ion range, the behaviour may be considered as similar to the impact of alpha particles, and of recoils of alpha decays and of neutrons.

In this study, we irradiated our specimens with ions of three different energies with GANIL facilities (Caen, France); thus we may note an eventual effect of the energy. In Table 1 we present the irradiations carried out: thanks to stopping powers we know that the major interactions at the beginning of the range are electronic, viz. inelastic collisions with target electrons, and that the nuclear collisions may be neglected.

Table 1 Spoiling of AlN by irradiations; R_p is the average projected range of the ions, $S_{e, ave}$ and $S_{n, ave}$ are the average electronic and nuclear stopping powers respectively along the first $5 \mu\text{m}$.

Ion irradiation	R_p (μm)	$S_{e, ave}$ (keV/nm)	$S_{n, ave}$ (keV/nm)
74 MeV Kr	9.7	10.9	0.05
92 MeV Xe	9.4	13.5	0.14
930 MeV Xe	46.2	22.1	0.01

The specimens were irradiated with fluences of 10^{12} , 10^{13} , 10^{14} , and 10^{15} ions per cm^2 , at room temperature (RT) and 773K for 74 MeV Kr, and only at RT for 92 MeV Xe, whereas with 930 MeV Xe they were irradiated with fluences of 10^{11} , 10^{12} , 10^{13} , and 4.5×10^{13} cm^{-2} at RT and 773K.

Characterization techniques

In order to know the defects induced by irradiations, thermally-stimulated luminescence (TSL) and optical absorption spectrophotometry (OAS) were used.

For TSL measurements, the samples were excited for five minutes with a deuterium lamp (50 W) of 200-400 nm spectral range. Then, two minutes after excitation, readout was performed during a linear heating of the sample from RT to 673K at a rate of $1 \text{ K} \cdot \text{s}^{-1}$. A photomultiplier (Philips 2018 B, 200-600 nm) was utilised to record the TSL intensity as a function of temperature, whereas spectral analysis was achieved with an optical multi-channel analyzer covering the 300-800 nm range.

OAS was performed at RT in the 250-800 nm range thanks to Varian Carry 500 spectrophotometer equipped with the diffuse reflection sphere (absorption reference: Teflon). Absorption bands affected by irradiation, were identified by subtracting the absorption spectrum of the virgin sample from the spectrum of each specimen after irradiation.

TSL and OAS allow us to identify defects in some microns near the surface, but the depth analysed is not easy to determine. That is why in Table 1 we calculated the average electronic and nuclear stopping powers on the first 5 μm , considering that the defects created at higher depth do not affect the TSL (because specimens are excited with UV radiations) and OAS signal.

Results and Discussion

Thermally-stimulated luminescence

TSL glow curves (Fig. 1.a) show one broad peak centered around 400 K, which attests the presence of trapping centers (TC), certainly oxygen in substitution of nitrogen O_N and/or bivalent 2O_N centers [15-17]. On the other hand, TSL spectra (Fig. 1.b) allow us to note 5 recombination centers (RC); the identification of the RC is shown in Table 2. The presence of chromium and manganese was confirmed by ICP: 1.9 and 2.0 ± 0.1 ppm, respectively.

Table 2 Identification of recombination centers in aluminum nitride [18, 19].

E (eV)	Recombination Center
1.79 ± 0.03	Cr_{Al}
2.04 ± 0.05	Mn_{Al}
2.67 ± 0.17	$\text{V}_{\text{Al}}-2\text{O}_\text{N}-2\text{N}$
3.17 ± 0.01	$\text{V}_{\text{Al}}-\text{O}_\text{N}-3\text{N}$
3.60 ± 0.04	Transition from V_{Al} to O_N

The main result is that whatever the irradiation is, no new TC nor RC is created. Nevertheless, the higher the fluence, the lower the TSL intensity.

According to the evolution of the TSL area as the function of fluence and temperature, a threshold electronic stopping power $S_{e, th}$ may be defined, the value of this threshold is between 14 and 22 keV nm^{-1} , the electronic stopping powers of 92 MeV Xe and 930 MeV Xe, respectively (Table 1). Thus, two behaviours are notable as the function of electronic stopping power. If $S_e < S_{e, th}$, the TSL area is similar

whatever the electronic stopping power is. Moreover, an annealing effect may be noticed for irradiation performed at 733K, inducing a less intense signal for the same fluences. On the other hand, if $S_e > S_{e, th}$, the TSL area decreases when S_e increases and no evident annealing effect can be noticed for 930 MeV Xe irradiation carried out at 773K.

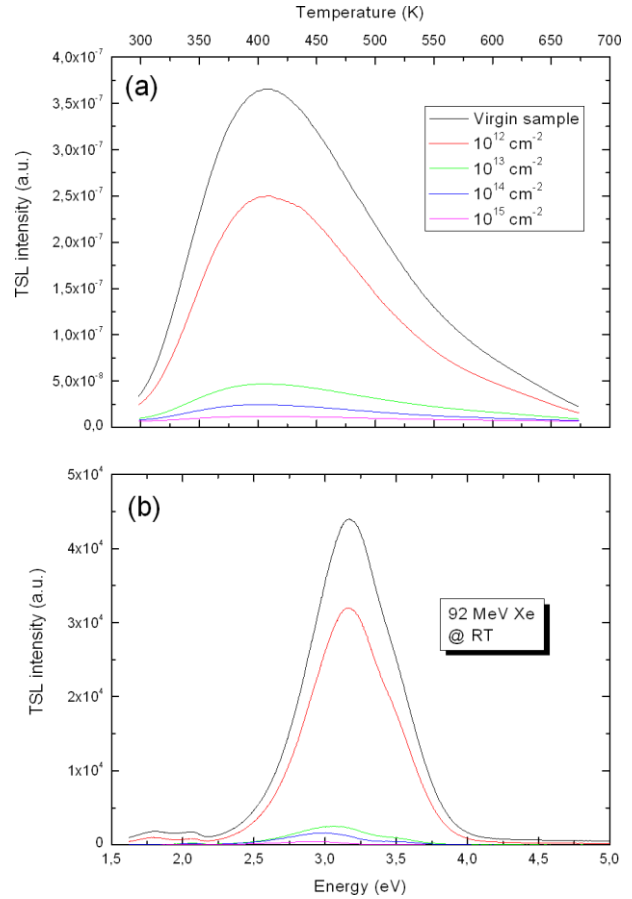


Fig. 1 TSL glow curves (a) and spectra at 400K (b) of aluminum nitride irradiated at room temperature with several fluences of 92 MeV Xe ions.

The decrease of the TSL area can be explained by three hypotheses. First, it can be the consequence of a decrease of the concentration of TC and/or RC. However, as irradiations create defects, this hypothesis seems not to be relevant. Second, there may be a creation of defects that absorb the TSL radiations. But, as the relative concentrations of RC seem to vary, this hypothesis seems not to be relevant either. Eventually, the third hypothesis would be a combination of both, with a variation of the concentration of TC and/or RC (decrease or increase) and an increase of the concentration of absorbing defects.

Optical absorption spectrophotometry

OAS was used in such a way to confirm the third hypothesis and to know the real effect of irradiations on TSL area.

OAS spectra allowed us to identify various absorption bands in UV range (Fig. 2.a) and in the visible range (Fig. 2.b), with certainly one band between both domains. The first important result is that no new

absorbing defect was detected after irradiations. However, some bands are affected, they are listed in Table 3.

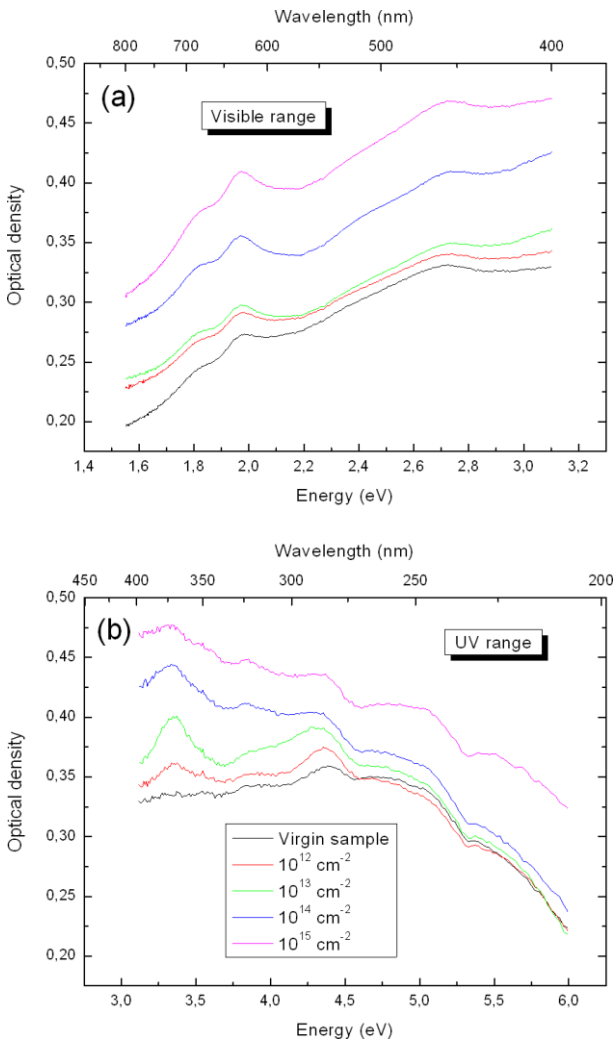


Fig. 2 Optical absorption of aluminum nitride in visible (a) and UV (b) domains as a function of fluence for irradiations carried out at room temperature with 92 MeV Xe ions.

Table 3 Identification of absorption bands affected by irradiations [20-24].

E (eV)	λ (nm)	Absorbing defect
1.83 ± 0.02	678 ± 7	Cr^{3+} ?
1.97 ± 0.02	630 ± 6	?
2.43 ± 0.03	511 ± 6	?
2.73 ± 0.05	455 ± 9	V_N or Al_i
3.06 ± 0.06	406 ± 8	Vacancy aggregates?
3.35 ± 0.05	370 ± 6	Vacancy aggregates?
4.28 ± 0.12	290 ± 8	O related

As the estimation of the concentration of each AD is complicated, we decided to follow the average absorption as a function of fluence for each irradiation. Thus, we noted two behaviours as a function of irradiation temperature. For irradiations carried out at RT, no effect of electronic stopping power was noted and the average absorption is the same for the three irradiations whatever the fluence is. On the other hand, if irradiation temperature is of 773K, electronic stopping

power has an effect: if $S_e < S_{e, th}$, we can observe an annealing effect; actually, absorption becomes less important than for the virgin sample, except for the highest studied fluence (10^{15} cm^{-2}). But if $S_e > S_{e, th}$, no annealing effect is noted except for the fluence higher than a critical fluence, which is in the 10^{13} - $4.5 \cdot 10^{13} \text{ cm}^{-2}$ range.

Conclusions

In this study, we attempted to identify, with thermally-stimulated luminescence and optical absorption spectrophotometry, the defects created by irradiation with swift heavy ions, simulating the impact of fission products. The first result is that whatever the irradiation is, no new defect detectable with both techniques was observed after irradiations.

Then, as both techniques complement each other, we were able to understand the effect of irradiation parameters on the overall concentration of TC and RC, and of AD. Thus, when the fluence increases, the concentration of TC and RC decreases, and the concentration of AD increases. Electronic stopping power has no effect on the AD concentration at RT; on the other hand, if $S_e > S_{e, th}$, the TC and RC concentration decreases when S_e increases. Eventually, the temperature does not change the concentration in TC and RC, but induce a decrease of AD concentration if $S_e > S_{e, th}$. But if $S_e > S_{e, th}$, AD concentration increases up to a critical fluence then stabilizes.

Acknowledgements

The authors would like to gratefully thank Isabelle Monnet (GANIL) for her great help during the irradiation of the samples, and Yves Jorand (INSA, Lyon, France) for providing the virgin materials. This study was partly funded by the French research group MATINEX.

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